Quantification of fugitive methane emissions from the biogas plant in Linköping (SE)

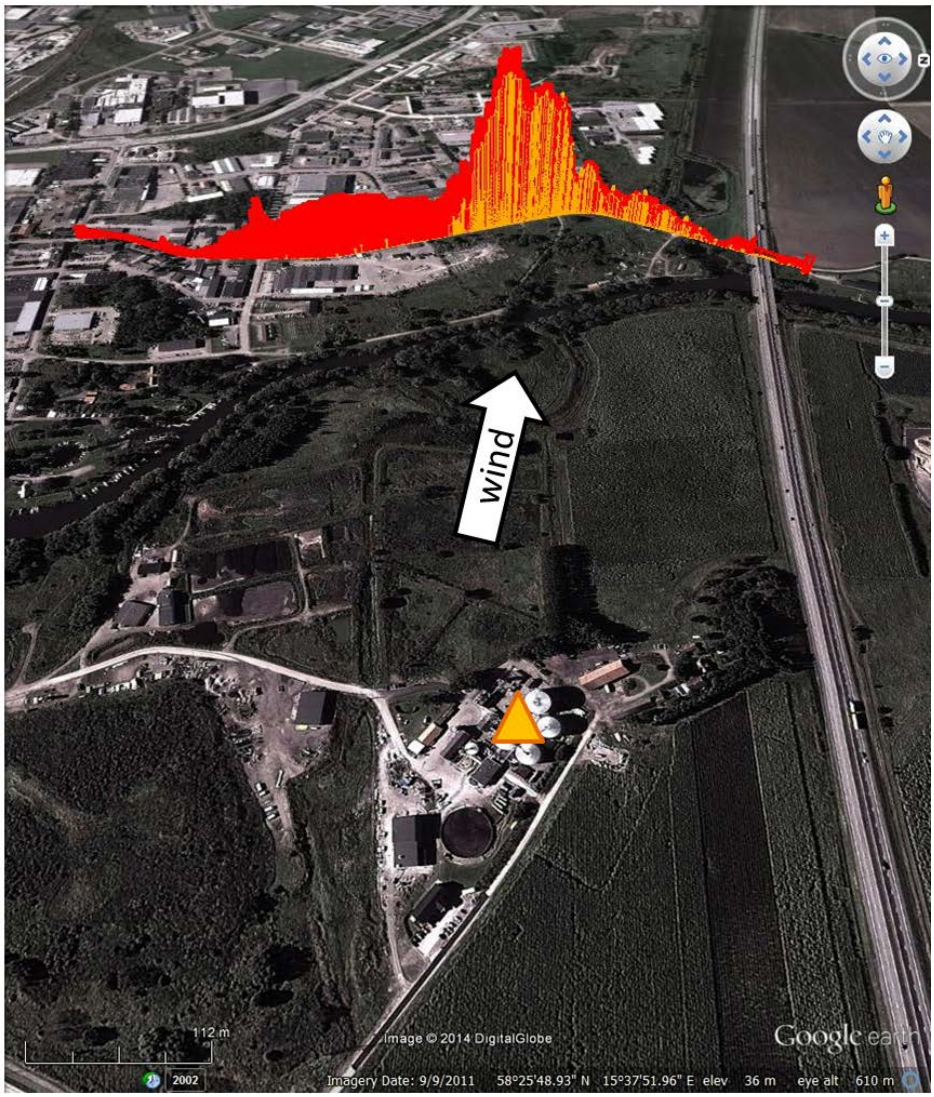
Delre, Antonio; Mønster, Jacob; Scheutz, Charlotte

Publication date:
2014

Document Version
Publisher's PDF, also known as Version of record

Technical University of Denmark

Quantification of fugitive methane emissions from the biogas plant in Linköping (SE)



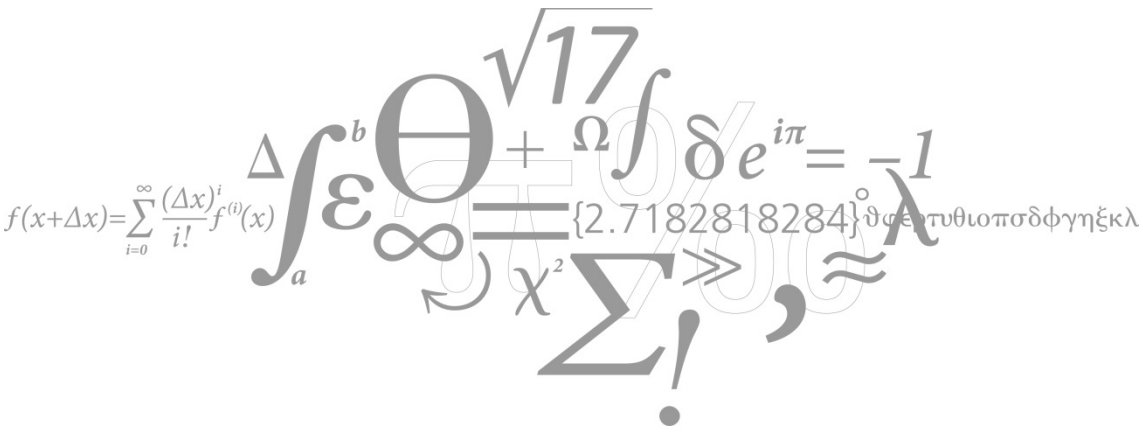
Atmospheric concentrations of methane and tracer gas above background level at approximately 900 meters downwind from the biogas plant measured on September 10th, 2014. The triangle marks the placement of the tracer gas release on the facility.

Antonio Delre, Jacob Mønster and Charlotte Scheutz

DTU Environment
Department of Environmental Engineering

October 2014

Contents



1. Introduction and purpose

Greenhouse gas (GHG) emissions from facilities treating organic waste are often difficult to quantify due to the diffusive nature of the emissions combined with large temporal variation and the challenging physical structure of the facility. Only over the last few years, the scientific community has developed methodologies and strategies of GHG quantifications from biogas facilities. However, there is no single measurement method that has been recognized as a standard method yet. The Technical University of Denmark has recently implemented a novel analytical setup enabling mobile measurements of small (ppb level) changes in atmospheric methane concentrations. This enables detection and quantification of methane emission sources by performing measurements downwind from the source in combination with release and measurement of a tracer gas. The analytical setup and the dynamic tracer dispersion method have been tested at a number of landfills and wastewater treatment plants since November 2011 (Mønster et al., 2014b; Yoshida et al., 2014), building up a sound knowledge on quantification of fugitive methane emissions from full-scale facilities.

The objective of this study was to quantify the methane emission from a Swedish biogas plant using the tracer dispersion method. The study was part of a large comparison study where other groups performed parallel methane detections and emission quantification using a range of different technologies including on-site measurements and remote sensing approach coupled to backward Lagrangian Stochastic inverse modelling.

2. Description of the measurement method

Total methane emissions were quantified using a mobile tracer dispersion method that combines a controlled release of tracer gas from the biogas facility with concentration measurements downwind of the facility, by using a mobile high-resolution analytical instrument (Mønster et al., 2014a; Yoshida et al., 2014).

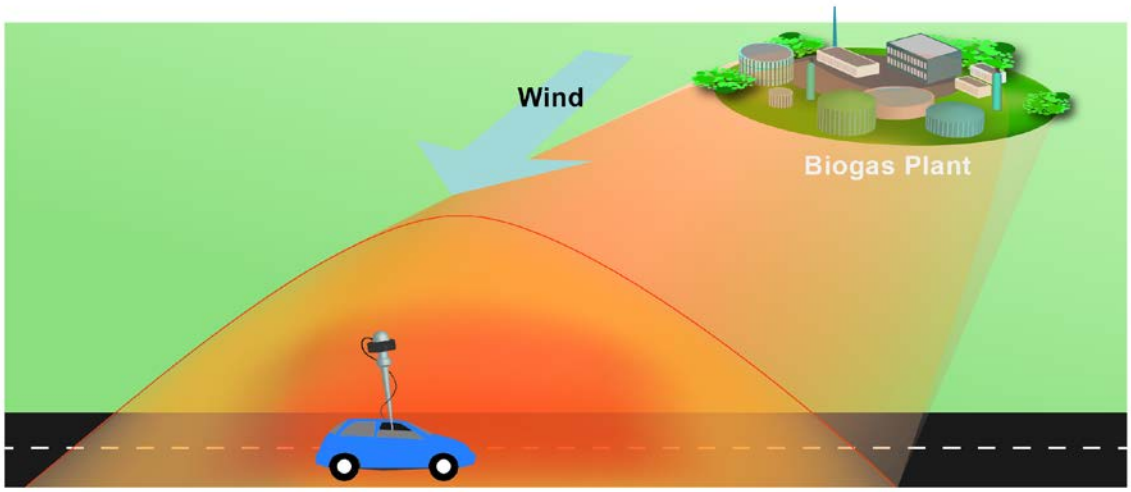


Figure 1. The principle of the dynamic plume method for quantifying GHG emissions from area sources

The tracer dispersion method, shown in Figure 1, is based on the principle that a tracer gas released at a source area, in this case a biogas facility, disperses into the atmosphere likewise the methane emitted from the same area. Since the ratio of their concentrations remains constant along their atmospheric dispersion, the methane emission rate can be calculated using the following expression when the tracer gas release rate is known:

$$E_{CH_4} = Q_{tr} * \frac{\int_{plume\ start}^{plume\ end}(C_{CH_4})dx}{\int_{plume\ start}^{plume\ end}(C_{tr})dx}\frac{MW_{CH_4}}{MW_{tr}}$$

where E_{CH_4} is the methane emission in mass per time, Q_{tr} is the tracer release in mass per time, C_{CH_4} and C_{tr} are the measured downwind concentrations in parts per billion (ppb) subtracted of their background concentrations and MW_{CH_4} and MW_{tr} are the molar weights of methane and tracer gas, respectively (Mønster et al., 2014a). In this study, acetylene (C_2H_2) was used as tracer due to its long atmospheric lifetime. Downwind plume concentrations were measured driving along transects with a cavity ring down spectrometer (CRDS) from Picarro (model G2203), which is a fast and high sensitive gas analyzer capable to detect methane and acetylene concentrations down to ppb level every second (Mønster et al, 2014a; Yoshida et al., 2014). A GPS was connected to the instrument for logging the measured concentrations to their geographical location. In order to obtain the best possible simulation of the source area, the tracer gas was released from the part of the plant where the most elevated methane concentration was seen and/or expected.

3. Description of the measurement campaign.

Measurements were performed from September 9th to September 12th, 2014. During the first two days, area and plant methane screenings were performed, followed by tracer release and methane emission quantification. The absence of favourable wind direction and speed during 11th and 12th allowed only a further confirmation of the plant screening. Different tracer gas release rates were tried out to have sufficient tracer gas for quantification in the downwind plume, but also to have tracer gas release for enough time to perform several plume traverses. Successful quantifications were done in the afternoon on Tuesday 9th and Wednesday 10th with a total tracer gas release of 0.44 kg h^{-1}. The tracer gas was released in one point from one gas bottle placed next to the gasholder in the digesters area. Figure 2 shows the securing of the acetylene cylinder, while Figure 3 shows its location.

Figure 2. Acetylene cylinder secured at the digesters area

Figure 3. Acetylene cylinder placement close to digester tanks marked with red circle

The measurements were performed during a period with stable weather conditions. On Tuesday 9th the sky was cloudy with light rain, the atmospheric pressure was around 1003 mbar and the air temperature was about 13 °C. On Wednesday 10th the sky was partly cloudy, the atmospheric pressure was around 1014 mbar and the air temperature was about 17 °C. On both days the wind blew from east with an average velocity of 1 m/s. While on Tuesday 9th twenty-one plume traverses were completed, on Wednesday 10th twenty-four plume traverses were carried out. The following two days the calm (no wind), sunny and warm weather resulted in a fast plume rising, which made methane measurements of downwind plumes at ground level far from the plant impossible.

4. Results and discussion

4.1 Biogas plant layout and gas production

The biogas plant in Linköping processes source separated household waste (≈50%), industrial food waste (≈25%), like dairy wastewater, and slaughterhouse waste (≈25%). After pre-treatment such as screening, grinding, watering and mixing, the substrate undergoes pasteurization followed by anaerobic digestion. The biogas plant has two digester tanks and one tank for biogas storage. 95% of the biogas is upgraded by a chemical scrubber, which sometimes is supported by a water scrubber that processes only about 5% of the upgraded biogas over one year. The digestate is stored in an open tank for 15-20 days before being delivered to farmers for land application. Figure 4 and Figure 5 give a visual overview of some of the process units. Notice that even though Figure 5 dates back to September 2011, it gives a very good understanding about the main units' locations in the plant.

Figure 4. Linköping biogas plant. From left to right: a) Food waste receiving area and pretreatment unit and thermal process tank, b) Main process units including digester tanks and biogas upgrading systems, and c. Digestate storage in open tank.

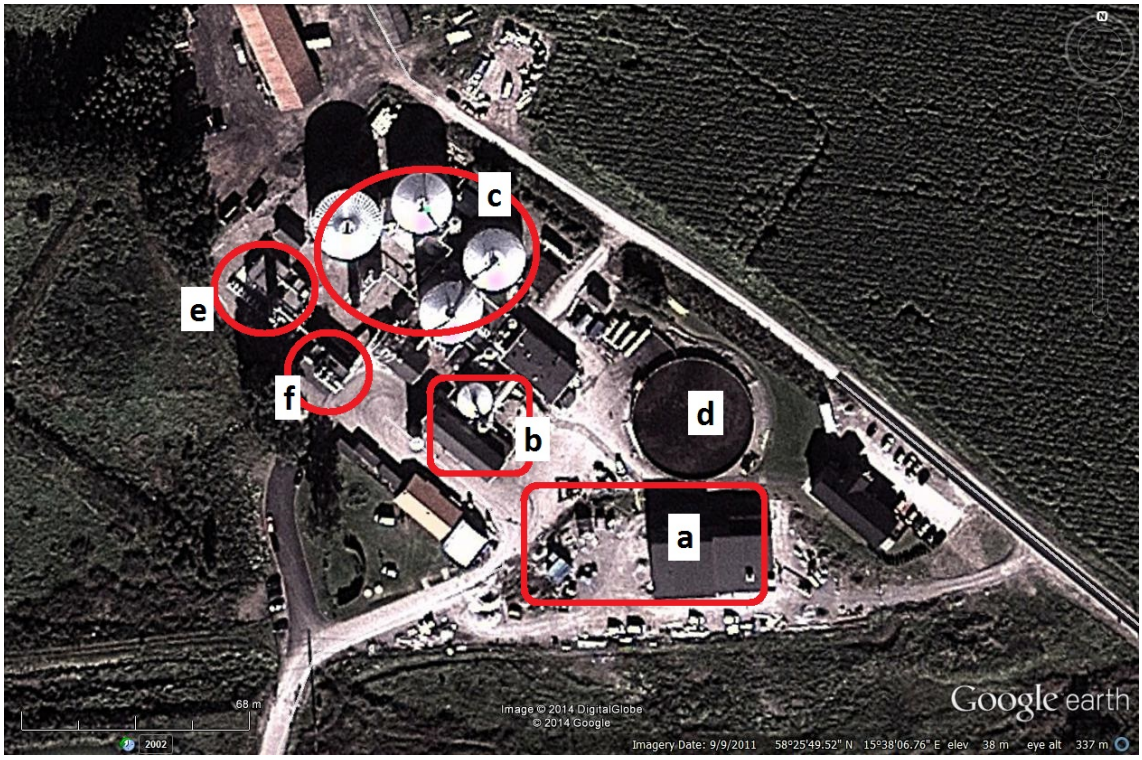


Figure 5 - Linköping biogas plant layout; a) Food waste pretreatment unit and thermal process tank, b) Substrate inlet and pasteurization tank, c) Digesters, gasholder and flare, d) Digestate storage in an open tank, e) Chemical scrubber and f) Water scrubber

Table 1 and Table 2 report biogas production during the days when the methane emissions quantifications were performed. Table 1 and Table 2 show the biogas and methane production before and after upgrading, respectively.

Table 1. Methane production before biogas upgrading process

Lapse of time	*Raw Biogas ($Nm^3\ h^{-1}$)*	*CH_4 content (%)*	*CH_4 production before upgrading ($Nm^3\ h^{-1}$)*
Tuesday 9th from 15:30 to 16:00	1910	60.0	1146
Wednesday 10th from 17:00 to 19:30	1800	63.0	1134

Table 2. Methane production after biogas upgrading process

Lapse of time	*Upgraded Biogas ($Nm^3\ h^{-1}$)*	*CH_4 content (%)*	*CH_4 production after upgrading ($Nm^3\ h^{-1}$)*
Tuesday 9th from 15:30 to 16:00	1180	96.2	1142
Wednesday 10th from 17:00 to 19:30	1184	96.0	1133

The volume of biogas is expressed according to standard temperature and pressure (STP) i.e. 0 °C and 1 atmosphere (DIN 1343). During measurements conducted on Wednesday 10th, biogas upgrading occurred with water scrubber support.

4.2 Methane screening of the area surrounding the biogas plant

Figure 6 shows different potential sources known to emit methane into the atmosphere in the surroundings of the biogas plant. North of the biogas plant is a sorting facility (receiving non-organic waste), while Northeast of the biogas plant is a landfill close to an incineration plant. The landfill is expected to release significant amounts of methane, whereas no methane emissions are expected from the incineration plant. South of the biogas plant is a wastewater treatment plant (WWTP), which stores its biosolids in heaps located relatively close to the biogas plant. The storage of biosolids could potentially emit methane.

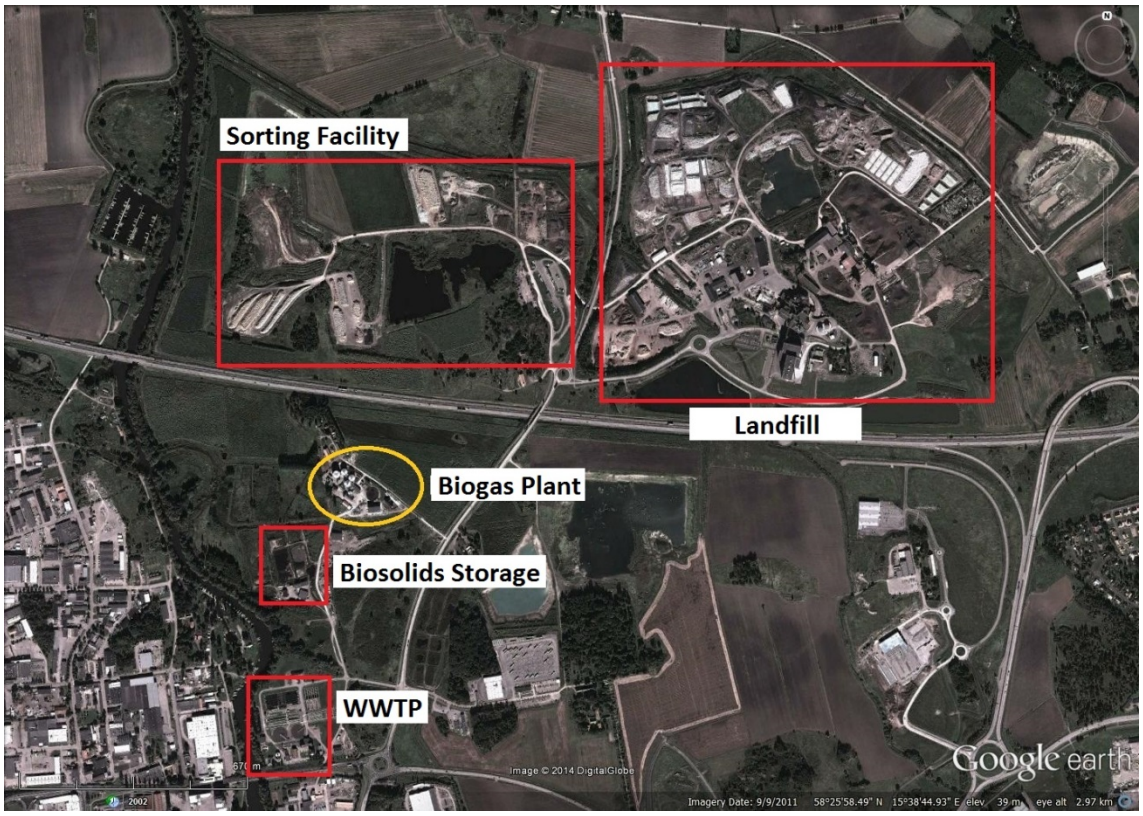


Figure 6. Location of treatment facilities in the surroundings of the biogas plant

The methane screening of the area is showed in Figure 7 where methane concentrations above background level are marked in red. Measurements were carried out with wind blowing from the East. Area screening upwind and downwind the biogas plant allowed distinguishing atmospheric methane plumes from different sources. Figure 7 shows methane concentrations measured at two different distances downwind the landfill. The lower concentrations measured at a further distance from the landfill underlines the atmospheric gas dispersion. Furthermore, methane plumes from the biogas plant, the biosolids storage and WWTP were observed. Notice that upwind of the three sources, the methane concentration was close to background. Emissions from the biogas plant are depicted in yellow. The distinction between emission from the WWTP biosolids storage and biogas plant was carried out making sure that the tracer release simulated the biogas plant well and that good mixing between methane and acetylene (C_2H_2) was obtained. Screening inside the sorting facility did not highlight any relevant methane releases.

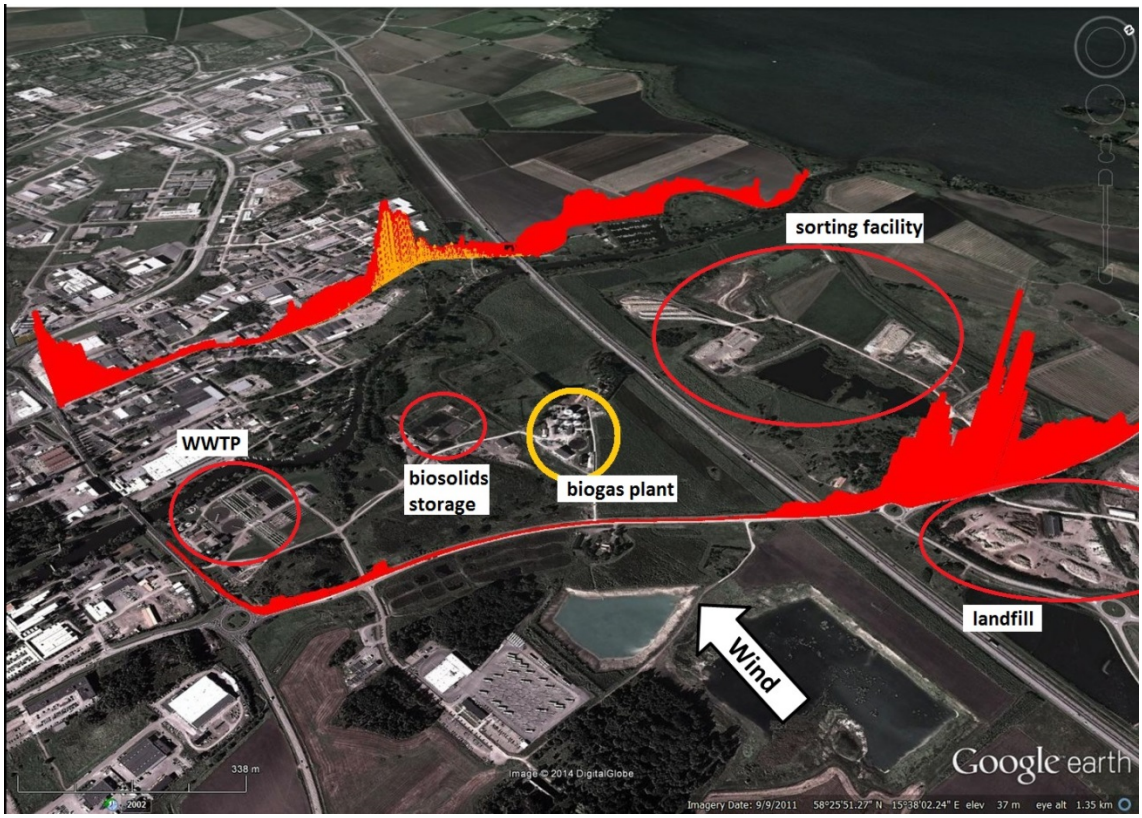


Figure 7. Methane screening of biogas plant surroundings with wind blowing from east.

4.3 Initial on-site methane screening of the biogas plant

A plant methane screening is usually used for identification of hotspots emission areas for optimal placement of the tracer gas cylinder to obtain the best methane source simulation. Methane might be released from the top of the digester tanks, which will be difficult to see based on on-site measurements due to the elevated release height in comparison to the measuring height (2m). Therefore in addition, information about the biogas plant and the methane plumes were used as support to identify the tracer gas placement.

Figure 8 shows methane concentrations measured during the plant screening at two different screening campaigns during calm and warm weather conditions with vertical plume rise from the area. Therefore, the detected methane concentrations are related to releases from the closest process units. The on-site screening indicated methane emissions from the open digestate storage tank, from the food waste pre-treatment area, from the biogas upgrading units and from the digester tanks.

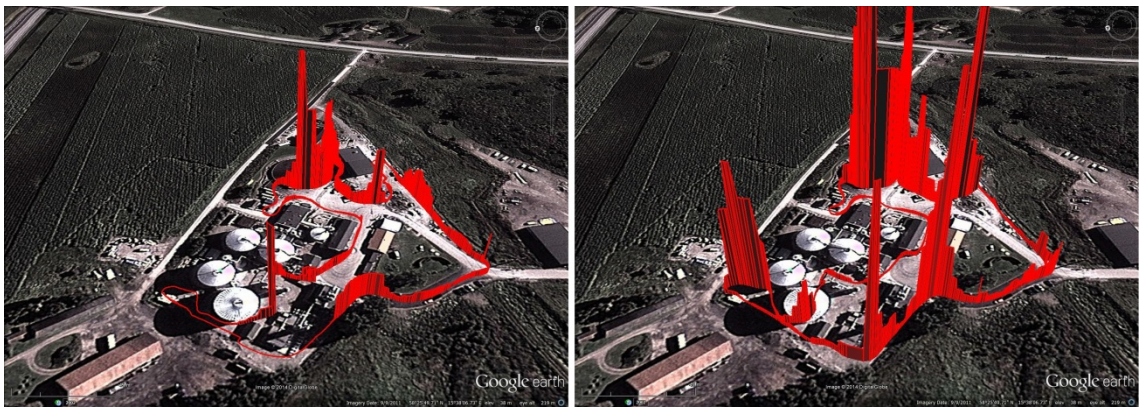


Figure 8. Biogas plant screening during calm and warm weather with plume vertical mixing. The maximum methane concentration above background was 9.0 ppm and 12.8 ppm during the two screenings, respectively.

4.4 Whole plant fugitive emissions

The placement of the tracer gas release was chosen in order to match biogas plant emissions and to distinguish methane released from the close WWTP biosolids storage. One tracer cylinder was chained up at the digesters area as reported in section 3. Figure 9 shows an example of downwind plumes, which was detected at 17:34 on Wednesday 10th along a road 800-900 m away from the plant, distant enough to consider the plant as point source. The tracer location is marked with a yellow triangle, while methane and acetylene plumes are showed in red and yellow, respectively. Peak concentrations above background level were 0.2 ppm of CH_4 and 3.1 ppb of C_2H_2. The tracer and the methane plume from the biogas plant followed each other nicely indicating a good simulation of the methane emission from the biogas plant. The figure also shows the adjacent methane plume coming from WWTP biosolids storage in southern direction. With wind from the East, it is possible to distinguish the two plumes from the biogas plant and the biosolids storage area from each other. However, if the wind shifts more to the North the plumes will blend together.

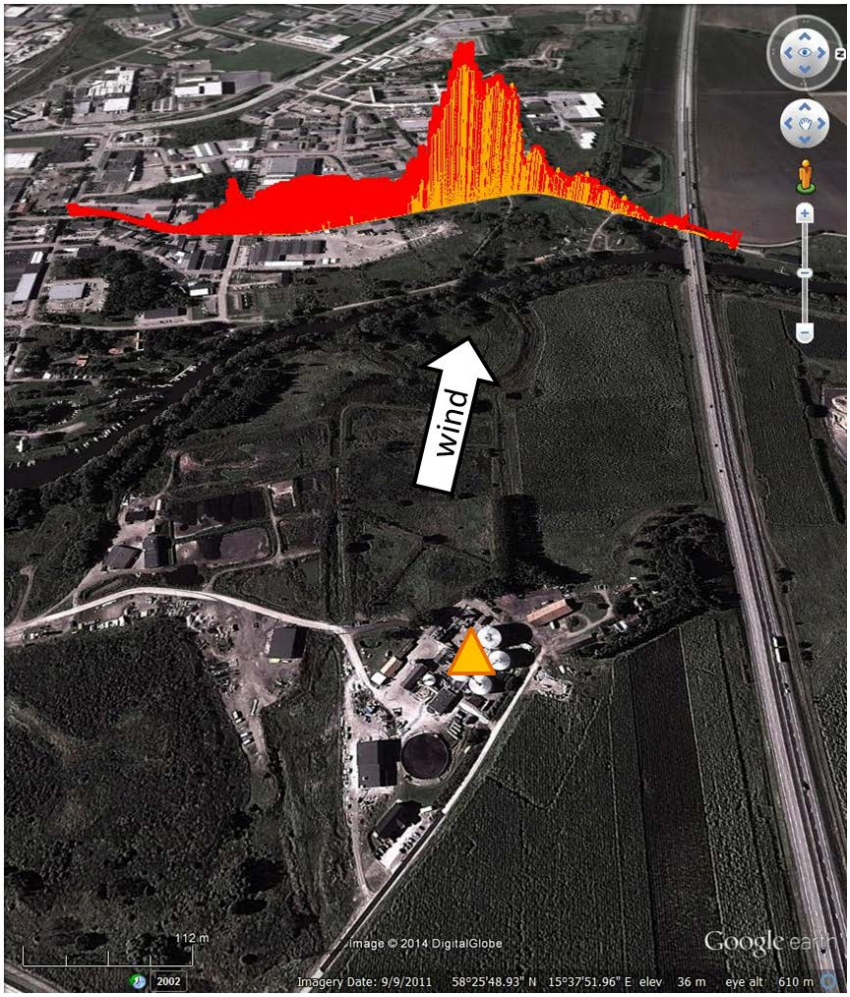


Figure 9. Atmospheric concentrations of methane and tracer gas above background level at approximately 900 meters downwind from the biogas plant measured on September 10th, 2014 at 17:34. The yellow triangle marks the placement of the C_2H_2 tracer gas release on the facility.

Further confirmation of correct tracer placement and emission source simulation is showed in Figure 10b by calculation of the coefficient of determination (R^2). The R^2 coefficient is obtained by plotting the plume concentrations of methane and acetylene to each other, and is an indicator of the degree of plume mixing. In this case, the R^2 was very high (0.94) indicating a good simulation of the methane plume.

For plume integration, the methane plumes form the biogas plant and the biosolids storage area have to be separated from each other. For most of the plume traverses, this was possible as there was very little overlap of the plumes. The methane plume separation was done using the tracer gas plume and tracking back the methane and tracer gas plume to the biogas plant using the wind direction. An example of methane plume separation is shown in Figure 10a. It is evident that the methane and the tracer gas plume from the biogas plant follows each other very nicely. However, it is also clear that there is another methane plume to the left, which comes from another source (the biosolids storage area) as there is no sign of the tracer. As the overlap of the plumes in this case was very little it is valid to separate the two plumes as shown in Figure 10a and b.

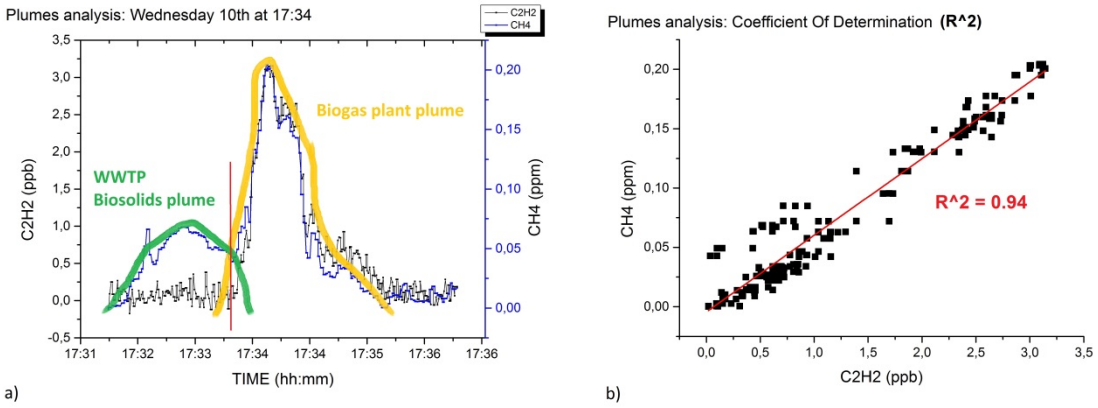


Figure 10. Data elaboration and confirmation of tracer placement; a) Concentrations over time b) Coefficient Of Determination (R^2) calculation.

Out of the more than forty plume transects performed on September 9th and 10th, 21 transects were useful for calculating the whole biogas plant methane emission. Plume transects where methane plumes were overlapping when the wind shifted more to North were thus not included. Single emission rates for each individual plume transects are listed in Table 3 and expressed both in kg CH_4 h^{-1} and Nm^3 CH_4 h^{-1} (STP: 1 atm, 0°C). Furthermore, emission factors (EF) related to raw biogas production and to upgraded biogas are reported. The EF is calculated as the ratio between the methane emission and the methane production for the specific measuring hours – both expressed in Nm^3 CH_4 h^{-1}.

The average methane emission rate from the biogas plant was found to be 23.6±1.8 kg CH_4 h^{-1}, which corresponds to 33.0±2.6 Nm^3 CH_4 h^{-1}. The confidence interval was calculated considering a t-distribution at significance level (α) of 5%, giving a confidence interval of 95% $(1-\alpha)$. This means that there is 95% probability that the right value falls within the confidence interval[1]. The smaller the confidence interval (*Conf.*) is, the higher is the accuracy of the methane quantification. In this case, the good accuracy was reached due to the high number of transverses included in the calculation (Mønster et al., 2014a). The plant emission factors, both referred to raw produced biogas (see Table 1) and upgraded biogas (see Table 2), were 2.9±0.2%, which is in the range of what was seen at other Swedish plants (Holmgren, 2014).

[1] The confidence interval was calculated as following: $\bar{y} \pm SEM * t_{\nu,\alpha/2}$

where $\bar{y}$ is the average value; *SEM* is the standard error of means ($SEM = SD/\sqrt{Count}$); *SD* is the standard deviation of the sample; *Count* is the number of transverses. $t_{\nu,\alpha/2}$ is the probability point of *t distribution* with ν degrees of freedom and significance level α as known as tail area probability. Therefore, such as confidence interval provides the range of values within the right value falls with $(1-\alpha)$ probability.

Table 3. Methane emission rates and Emission Factors (EF) measured at the biogas plant.

Day	*Peak time*	*Emission*	*Emission*	*EF Raw biogas*	*EF Upgraded biogas*
	(hh:mm)	*(kg CH_4 h^{-1})*	*(Nm^3 CH_4 h^{-1})*	*(%)*	*(%)*
Tuesday 9th	15:36	20.8	29.1	2.5	2.6
	15:46	18.4	25.8	2.3	2.3
	15:59	14.6	20.4	1.8	1.8
Wednesday 10th	17:11	22.6	31.7	2.8	2.8
	17:22	22.7	31.8	2.8	2.8
	17:26	24.8	34.7	3.1	3.1
	17:34	16.6	23.2	2.0	2.0
	17:40	19.6	27.5	2.4	2.4
	17:49	23.2	32.5	2.9	2.9
	18:01	25.9	36.3	3.2	3.2
	18:06	26.9	37.6	3.3	3.3
	18:33	28.4	39.7	3.5	3.5
	18:39	20.2	28.3	2.5	2.5
	18:44	23.7	33.2	2.9	2.9
	18:50	26.0	36.4	3.2	3.2
	18:55	26.0	36.4	3.2	3.2
	19:03	24.7	34.6	3.0	3.1
	19:06	29.8	41.7	3.7	3.7
	19:09	25.9	36.3	3.2	3.2
	19:16	25.3	35.4	3.1	3.1
	19:22	29.4	41.2	3.6	3.6
Average		***23.6***	***33.0***	***2.9***	***2.9***
Conf.*		***1.8***	***2.6***	***0.2***	***0.2***
SD		4.1	5.7	0.5	0.5
Count		21	21	21	21
SEM		0.9	1.2	0.1	0.1

Conf.: Confidence Interval calculated with t distribution at significance level α=5%

SD: standard deviation, Count: number of transects (dimensionless value), SEM: standard error of means.

Using information about the plant layout and the wind direction during measurements, quantification of emissions coming from digesters and gasholder area could be estimated. Wind blowing from East, as showed in. Figure 11, generates a downwind plume whose northern part (marked in green) describes emissions coming from digesters and gasholder area, whereas the southern part (marked in light blue) shows emissions coming from other process units.

Therefore, the calculation for emissions generated from digesters and gasholder area was carried out integrating only the green part of the acetylene and methane plumes.

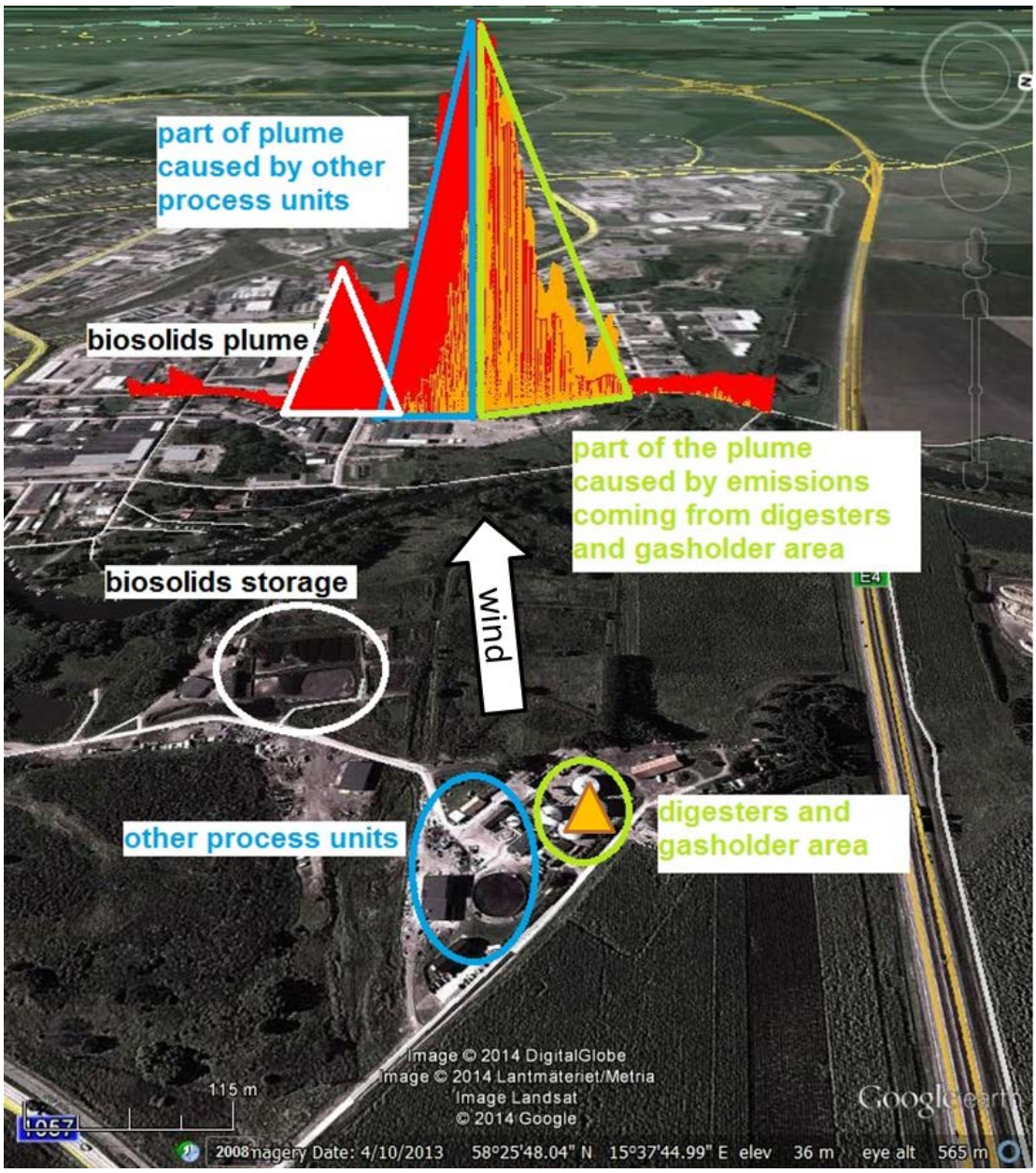


Figure 11. Estimation of methane emissions from digesters and gasholder area. Only the green part of the plume was used for this calculation. Downwind plume measured on September 10th, 2014 at 18:49.

Table 4 lists the methane emission estimated from the area with the digesters and the gasholder for each successful transect. The values are compared with the whole plant methane emissions in order to estimate the percentage of methane emissions coming from this area. The average emission from the area with the digester tanks and the gasholder was 14.9±0.9 kg CH_4 h^{-1}, which corresponds to 65±6% of the total emission of the biogas plant. The standard deviation of 14% (see Table 4) underlines the high uncertainty of this value, which therefore can be used only for a rough estimation.

Table 4. Estimation of CH_4 fugitive emissions from digesters area

Day of September 2014	*Peak time (hh:mm)*	*Whole plant Emission (kg CH_4 h^{-1})*	*Emission estimation from Digesters Area (kg CH_4 h^{-1})*	*Emission share from Digesters Area (%)*
Tuesday 9th	15:36	20.8	18.1	87
	15:46	18.4	15.4	83
	15:59	14.6	11.7	80
Wednesday 10th	17:11	22.6	16.2	72
	17:22	22.7	16.7	74
	17:26	24.8	17.8	72
	17:34	16.6	14.4	87
	17:40	19.6	15.5	79
	17:49	23.2	14.7	63
	18:01	25.9	16.2	62
	18:06	26.9	13.3	49
	18:33	28.4	13.5	48
	18:39	20.2	10.5	52
	18:44	23.7	17.0	72
	18:50	26.0	17.8	68
	18:55	26.0	12.4	48
	19:03	24.7	14.5	59
	19:06	29.8	13.5	45
	19:09	25.9	15.3	59
	19:16	25.3	13.7	54
	19:22	29.4	14.3	49
Average		***23.6***	***14.9***	***65***
Conf.		***1.8***	***0.9***	***6***
SD		4.1	2.0	14
Count		21	21	21
SEM		0.88	0.45	3

Conf.: Confidence Interval calculated with t distribution at significance level α=5%

SD: standard deviation, Count: number of transects (dimensionless value), SEM: standard error of means.

More information can be drawn comparing results from different days. Table 5 reports these calculations from data listed in Table 3. The average emission rate measured on Tuesday 9th was 17.9±7.8 kg CH_4 h^{-1}, which corresponds to 25.1±10.9 Nm^3 CH_4 h^{-1}, and on Wednesday 10th 24.5±1.7 kg CH_4 h^{-1}, which corresponds to 34.4±2.4 Nm^3 CH_4 h^{-1}.

EFs calculated for data collected on Tuesday 9th are 0.8% smaller than those calculated for data collected on Wednesday 10th. The different number of transects between the two quantification days, 3 on 9th vs 18 on 10th, involves different confidence interval underlining a more accurate value on Wednesday (0.2%) than Tuesday (1.0%). Nevertheless, the higher fugitive methane emissions on Wednesday 10th compared to Tuesday 9th could be explained by the additional use of water scrubber in the second day (see introduction to section 4), which is a technology known to release more CH_4 to the atmosphere than chemical scrubber (Petersson, 2012). Notice that also in this case the two EFs have the same value.

Table 5. Whole plant fugitive emissions in two different measurements days

	Whole plant emissions (kg CH_4 h^{-1})		*Whole plant emissions (Nm^3 h^{-1})*		*Emission Factor (%) Raw biogas*		*Emission Factor (%) Upgraded biogas*	
Day of sep-14	*Tue 9th*	*Wed 10th*	*Tue 9th*	*Wed 10th*	*Tue 9th*	*Wed 10th*	*Tue 9th*	*Wed 10th*
Average	***17.9***	***24.5***	***25.1***	***34.4***	***2.2***	***3.0***	***2.2***	***3.0***
Conf.	***7.8***	***1.7***	***10.9***	***2.4***	***1.0***	***0.2***	***1.0***	***0.2***
SD	3.1	3.4	4.4	4.8	0.4	0.4	0.4	0.4
Count	3	18	3	18	3	18	3	18
SEM	1.8	0.8	2.5	1.1	0.2	0.1	0.2	0.1

Conf.: Confidence Interval calculated with t distribution at significance level α=5%

SD: standard deviation, Count: number of transects (dimensionless value), SEM: standard error of means.

5. Conclusion

Off-site and on-site methane screenings indicated methane releases from digesters, biogas upgrading units, digestate storage tank and pre-treatment area. The methane emission from Linköping biogas plant, during the afternoons of September 9th and September 10th, was successfully quantified using the tracer dispersion method. The average emission rate measured was 23.6±1.8 kg CH_4 h^{-1} (corresponding to 33.0±2.6 Nm^3 CH_4 h^{-1}). Plant emission factors (EFs) referred to raw produced biogas and upgraded biogas were 2.9±0.2%. The methane emission from the area with the digester tanks were estimated to account for approximately 65±6% of the total emission from the plant. The remaining part of the methane emission came from the pre-treatment area and the digestate storage in an open tank.

On Wednesday 10th the methane emission was higher (24.5±1.7 kg CH_4 h^{-1}, which corresponds to 34.4±2.4 Nm^3 CH_4 h^{-1}) than the emission measured on Tuesday 9th (17.9±7.8 kg CH_4 h^{-1}, which corresponds to 25.1±10.9 Nm^3 CH_4 h^{-1}). This might be explained by the additional use of water scrubber during the second day, which is a technology known to release more CH_4 in the atmosphere than chemical scrubber.

Stable weather conditions characterized each methane quantification day during measurements. In both days, wind blew from East with an average velocity of 1 m/s. The following two days (September 11th and 12th) the calm, sunny and warm weather resulted in a fast plume rising, which made impossible methane measurements of downwind plumes at ground level far from the plant impossible.

6. References

Holmgren, M.A., 2014. Methane Emission from Swedish Biogas Plants, in: 2nd IBBA Workshop on Methane Emissions in Kiel, Germany, September 4th 2014. SCG - Swedish Gas and Technology Center.

Mønster, J.G., Samuelsson, J., Kjeldsen, P., Rella, C.W., Scheutz, C., 2014a. Quantifying methane emission from fugitive sources by combining tracer release and downwind

measurements - a sensitivity analysis based on multiple field surveys. Waste Manag. 34, 1416–28.

Mønster, J., Samuelsson, J., Kjeldsen, P., Scheutz, C. 2014b. Quantification of methane emission from 15 Danish landfills using mobile tracer dispersion method. Waste Management. *In press.*

Petersson, A., 2012. The Swedish Voluntary system for control of methane emissions.

Yoshida, H., Mønster, J., Scheutz, C., 2014. Plant-integrated measurement of greenhouse gas emissions from a municipal wastewater treatment plant. Water Res. 1, 108–118.